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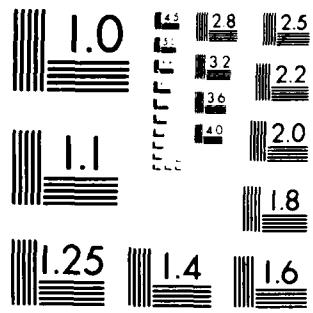
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A MODEL FOR THE INTERACTION OF TWO ELECTRIC DOUBLE LAYERS
IN TWO DIMENSIONS: THE METAL ELECTROLYTE INTERFACE, AND THE
THE DONNAN MEMBRANE

by

L. BLUM

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Department of Physics, POB AT, Faculty of Natural Sciences, University of
Puerto Rico, Río Piedras, Puerto Rico 00931

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A second case of interest is the case of the two media separated by a gap that in our case could be charged: this is a model of a two dimensional membrane or the polarization of the inner Helmholtz region. Very surprisingly, we find that also the surface dipole is completely screened by the charges surrounding the interface, so that the potential drop across the interface layer only depends on the logarithm of the charge densities, as found by Ballone, Senatore and Tosi for the most simple case of a discharged contact.



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*
L.Blum

Department of Physics,POB AT, Faculty of Natural Sciences, University of
Puerto Rico,Rio Piedras,Puerto Rico 00931.

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ABSTRACT

A model for the interface of two media with different background densities , separated by a charged bilayer, is solved exactly in a two dimensional, one component plasma at reduced temperature 2.

when the two media are in direct contact (no gap), then we can think of it as a model of a classical metal and an electrolytic solution or just two metals. The contact potential, or potential of zero charge appears to be a very simple function of the ratio of the background densities we also find that the potential does not depend on the surface charge,a fact that we explain using a corollary of the perfect screening theorem.

A second case of interest is tha case of the two media separated by a gap that in our case could be charged :this is a model of a two dimensional membrane,or the polarization layer of the inner Helmholtz region. Very surprisingly,we find that also the surface dipole is completely screened by the charges surrounding the interface,so that the potential drop across the interface only depends on the logarithm of the charge densities, as found by Ballone,Senatore and Fosi for the most simple case of a discharged contact.

I-INTRODUCTION

One of the most interesting exact results in the theory of charged interfaces in recent times is the solution of the one component plasma (OCP) in two dimensions by Jancovici (1,2). Although this model is exactly solvable only at reduced temperature 2, which in a three dimensional world corresponds to a little too concentrated ionic solution (for biological systems), it may be solved for a rather wide variety of inhomogeneous systems. One of these is the interface between two media of different background density, separated by a charged gap. This case could be a model for the classical metal-metal junction, the semiconductor junction, the metal electrolyte interface among others.

In Biology there is also a system which corresponds to this model: The Donnan equilibrium is established between two media containing different concentrations of proteins, and which are separated by a membrane that allows passage of the small ions only (3). A system of particular interest is the nerve membrane (4,5). The mechanism of production of the so called action potential is strongly related to the charge distribution across the interfaces. Because of the small size of the system and the low conductivity, direct electrochemical measurements are difficult. Only recently experiments involving interacting double layers in a system similar to a Donnan system have been reported (6). Here two immiscible electrolytes are studied electrochemically. However these experiments involve rather complex organic molecules, and it is very hard to construct a microscopically correct theory for the charge and potential profiles, and the differential capacitances. We should mention in this context the recent work of Levine and Cuthwaite (7), Nicollis and Pratt (8) and Allastuey and Levesque (9).

The purpose of this work is to present a simple model for these systems based on the two dimensional OCP. Forrester and Smith(10) have solved the case of a plasma contained between two fixed walls, and Smith solved the case of a flat ideal electrode with image forces (11). Our method of solution is based on the method of Jancovici (1,2), but follows the outline of these last two references. It is our hope that this exact solution will serve as a benchmark to evaluate the accuracy of different approximate theories, and to check the validity of exact theorems such as the perfect screening sum rules.

In section 2 we give a description of the model and a brief outline of the method of solution. The reader interested in the technical details of the solution should consult references (1,2,10,11). In section 3 we discuss the case in which the width of the gap is zero. In section 4 we present calculations for the case in which the membrane, or gap, is of finite width. We remark that this case also represents the inner Helmholtz layer with a fixed dipole.

II-METHOD OF SOLUTION

The system consists of an empty strip of width ℓ , that separates the two plasmas. The neutralizing background in each side is of density $-e\alpha/\Gamma_1$ and $-e\alpha/\Gamma_2$, where e is the elementary charge, and the borders of the strip have charge densities $-e\sigma_1/\Gamma_1$ and $-e\sigma_2/\Gamma_2$

The plasma coupling parameter is

$$\Gamma = \beta e^2 / \epsilon = 2$$

where $\beta = 1/kT$ is the Boltzmann thermal factor, T being the absolute temperature. The value $\Gamma = 2$ may be special in the sense that the pair correlations of the bulk have gaussian rather than exponential screening. Otherwise this parameter is not unphysical, and, in fact it corresponds to the order of magnitude of a 1M electrolytic solution at room temperature.

Following Jancovici (1,2) we consider initially our system to be confined to a disk of radius R . In this disk there is a ring of inner radius R_1 and outer radius R_2 . The uniform background density of the inner region is $-e\alpha/\Gamma_1$, and that of the outer region is $-e\alpha/\Gamma_2$

Clearly

$$R_2 = R_1 + \ell \quad (2.1)$$

where ℓ is the width of the ring which eventually will become our membrane. In our model this membrane will be just a gap. The mobile ions of charge e are free to be anywhere.

The total number of ions N must satisfy the electroneutrality relation

$$N = 2(\sigma_{11}^2 R_1 + \sigma_{22}^2 R_2) + R_1 \alpha_{11}^2 + (R_2 - R_1) \alpha_{12}^2 \quad (2.2)$$

The hamiltonian of this model is

$$\begin{aligned} H = & (1/2)e \left\{ - \sum_{k>j}^N \frac{1}{r_{kj}} \ln|r_{kj}|^2 + \sum_{k=1}^N [2\sigma_{11}^2 R_1 \ln(r_{k1}/R_1)^2 \Theta(r_{k1}-R_1) \right. \\ & 2\sigma_{22}^2 R_2 \ln(r_{k2}/R_2)^2 \Theta(r_{k2}-R_2) + \alpha_{11}^2 (r_{k1}-R_1)^2 \Theta(R_1-r_{k1}) \\ & \left. + \alpha_{12}^2 R_1 \ln(r_{k1}/R_1)^2 \Theta(r_{k1}-R_1) \right. \\ & \left. + \alpha_{22}^2 (r_{k2}-R_2)^2 - \alpha_{12}^2 R_1 \ln(r_{k2}/R_2)^2 \Theta(r_{k2}-R_2) \right] \right\} + \frac{\beta}{N} \quad (2.3) \end{aligned}$$

where r_k is the position of ion k , of charge e , and $\Theta(x)$ is the Heaviside function of x . β is a background term that is irrelevant to our present

calculation. The potentials have been shifted to insure continuity across the plates.

We compute the canonical partition function

$$Z = (1/N!) \int d^N r e^{-\beta H} \quad (2.4)$$

Replacing (2.3) into (2.4)

$$Z = e^{-\beta N!} (2\pi)^N \prod_{n=0}^{N-1} \left\{ \int_0^{\infty} dr_n n!^{-1} e^{-\beta r_n^2} \right\}^{\infty} \frac{-\alpha_{12}^2 (r_{k1}-R_1)^2}{e^{2\beta R_1}}$$

$$\begin{aligned}
 & + [1/R_1^{g_1}] \int_{R_1}^{\infty} dr r^{2n+1} \\
 & + [1/R_1^{g_1}][1/R_2^{g_2}] \int_{R_2}^{\infty} dr r^{2n+1} e^{-\alpha_2^2(r^2 - R_2^2)} \quad \{ \\
 & \quad (2.5)
 \end{aligned}$$

where

$$n = n - 2\sigma_1^2 R_1 - \alpha_1^2 R_1^2 \quad (2.6)$$

$$n = n - 2\sigma_1^2 R_1 - 2\sigma_2^2 R_2 + (\alpha_2^2 R_2^2 - \alpha_1^2 R_1^2) \quad (2.7)$$

and $\beta = (1/kT)$ is the Boltzmann factor.

Introducing the incomplete gamma function

$$\gamma(a, b) = \int_0^b dt t^{a-1} e^{-t} \quad (2.8)$$

we get

$$Z = e^{-\beta B} \left(\prod_{n=0}^N \Gamma \right)^N \{ \gamma(n+1; \frac{\alpha_1^2 R_1^2}{1}, \frac{\alpha_2^2 R_2^2}{1}) \exp[\alpha_1^2 R_1^2]/\alpha_1^{n+1}$$

$$\begin{aligned}
& + \left[\frac{1}{R_1} \right] \left[\frac{1/2(n+1)}{g_2} \right] \left[\frac{R_2}{g_1} \right] \left[\frac{-R_1}{g_2} \right] + \\
& + \left[\frac{1/R_1}{g_2} \right] \left[\frac{1/R_2}{g_2} \right] \left[\frac{\gamma_{22}(n+1; \alpha R_1^2)}{2} - \frac{\gamma_{22}(n+1; \alpha R_2^2)}{2} \right] + \\
& \exp \left[\frac{\alpha R_1^2}{2} \right] \left(\frac{1/\alpha}{2} \right)^{n+1}
\end{aligned} \tag{2.9}$$

The single particle density is

$$\begin{aligned}
\rho(r) = & \sum_{n=0}^{N-1} \frac{(1/D_n)}{n} \left\{ \exp \left[-\frac{\alpha}{2} \left(\frac{r-R_1}{2} \right)^2 \right] \left(\frac{r/R_1}{2} \right)^{2n} \Theta \left(\frac{R_1 - r}{2} \right) + \right. \\
& \left. \left(\frac{r/R_1}{2} \right)^{2n} \left[\Theta \left(\frac{r - R_1}{2} \right) - \Theta \left(\frac{r - R_2}{2} \right) \right] + \right. \\
& \left. + \left(\frac{r/R_2}{2} \right)^{2n} \left(\frac{R_2/R_1}{2} \right)^q \exp \left[-\frac{\alpha}{2} \left(\frac{r - R_2}{2} \right)^2 \right] \left[\Theta \left(\frac{r - R_2}{2} \right) \right] \right\}
\end{aligned} \tag{2.10}$$

with

$$D_n = \exp \left(\frac{\alpha R_1^2}{2} \right) \left(\frac{1/R_1}{2} \right)^{2n} \gamma_{22}(n+1; \alpha R_1^2) +$$

$$+ \left(\frac{r}{2} / (2n+2) \right)^{2n+2} \left[\left(\frac{R_2/R_1}{2} \right)^q - 1 \right]$$

$$\begin{aligned}
& + \left(\frac{1}{R_1} \right)^{2n} \left(\frac{1}{R_2} \right)^{2(n-n)} [\delta_{21}^{(n+1; R_1)} - \delta_{22}^{(n+1; R_2)}] \\
& \exp[\alpha_2^2 R_2^2] / \alpha_2^{n+1}
\end{aligned} \tag{2.11}$$

Similarly, the pair density distribution function is

$$\begin{aligned}
\rho(r_1, r_2) &= \rho(r_1) \rho(r_2) - \sum_{m_1, m_2}^{m_1, m_2} r_1^{m_1} \left(\frac{r_1}{R_1} \right)^{2m_1} r_2^{m_2} \left(\frac{r_2}{R_2} \right)^{2m_2} \left[\frac{1}{R_1} \right]^{2(m_1+m_2)} \\
& \{ G(r_1) G(r_2) \} / \{ D_{m_1, m_2} \}
\end{aligned} \tag{2.12}$$

where D_m is defined in (2.11) and $G(r)$ is given by

$$\begin{aligned}
G(r) &= \exp[-\alpha_1^2 (r_1^2 - R_1^2)] \Theta(R_1 - r_1) \\
& + (r_1/R_1)^2 \Delta_g \\
& + (r_2/R_2)^2 \Delta_2 \\
& + (R_2/R_1)^2 (r_2/R_2)^2 \exp[-\alpha_2^2 (r_2^2 - R_2^2)] \Theta(R_2 - r_2)
\end{aligned}$$

(2.13)

$$\text{with } \frac{\Delta}{g} = n - n$$

$$\frac{\Delta}{2} = n - n$$

In the limit

$$R_1, R_2 \rightarrow \infty$$

$$\frac{R_2 - R_1}{2} = \ell \quad (2.14)$$

and using

$$\delta(n+1; N) = (\sqrt{2\pi} n/2) \exp(-n+n \ln n) \left\{ 1 + \frac{1}{2} [(N-n)/\sqrt{2n}] \right\}$$

with

$$\frac{1}{2}(x) = (2/\sqrt{\pi}) \int_0^x dt e^{-t^2} \quad (2.15)$$

we get

$$P(x) = (2/\sqrt{\pi}) \int_{-\infty}^{\infty} dt \frac{(1/D)}{t} \left\{ \exp\left(-2\alpha \frac{x^2}{t} - 2xt\sqrt{\sigma}\right) \Theta(-x) + \right.$$

$$\left. \exp\left(-2x\sqrt{2}(t+2\sigma)\right) [\Theta(x)-\Theta(x-\ell)] \right\}$$

$$+ \exp\left[-2\alpha \frac{(x-\ell)^2}{t} - 2(x-\ell)\sqrt{2(t+\sqrt{2}\sigma+\sqrt{2}\sigma)} - 2\ell(2\sigma+\sqrt{2}t)\right] \Theta(x-\ell) \quad (2.16)$$

with

$$\begin{aligned} D &= \exp(t/\alpha) \left[1 + \frac{\int_1^t (\tau/\sqrt{\alpha})}{\sqrt{\alpha}} \right] / \sqrt{\alpha} \\ &- [\exp\{-2\int_1^t \sqrt{2}(t+\sqrt{2}\sigma_1)\} - 1] / [\int_1^t \sqrt{1+(2\sigma_1)^2}] \\ &+ \exp\{-2\int_1^t \sqrt{2}(t+\sqrt{2}\sigma_1) + [t+\sqrt{2}(\sigma_1 + \sigma_2)]^2/2\} \left[1 - \frac{\int_1^t (t+\sqrt{2}(\sigma_1 + \sigma_2)/\sqrt{\alpha})}{\sqrt{\alpha}} \right] / \sqrt{\alpha} \end{aligned} \quad (2.17)$$

and for the pair density

$$\rho(z_1, z_2) = \rho(x_1) \rho(x_2) - \exp[-2\alpha x_i^2 - 2\alpha x_j^2]$$
$$4/\pi^3 \left| \int_{-\infty}^{\infty} dt \exp[-2t(x_1 + x_2 - iy)] / D_t \right|^2 \quad (2.18)$$

where $z = x + iy$, a result due to Jancovici (private communication, 12).

III THE BACKGROUND JUMP MODEL

A very interesting particular case of (2.16) arises when the width of the interface layer ℓ is zero. In that case the charge density is

$$\rho(x) = \frac{(2/\sqrt{\pi})}{t} \int_{-\infty}^{\infty} \frac{(1/D)}{t} \left\{ \exp\left(-2\alpha x - \frac{2}{t}\right) \right\} \quad x < 0$$

$$\sigma(x) = \frac{(2/\sqrt{\pi})}{t} \int_{-\infty}^{\infty} \frac{(1/D)}{t} \exp\left(-2\alpha x - \frac{2x\sqrt{2}(t+\sqrt{2}\sigma)}{t}\right) \quad x > 0$$
(3.1)

$$D = \frac{1}{t} \exp\left(\frac{t}{\alpha}\right) [1 + \frac{\Phi(t/\sqrt{\alpha})}{\sqrt{\alpha}}]$$

$$+ \exp\left(\frac{(t+\sqrt{2}\sigma)}{\alpha}\right) [1 - \frac{\Phi(t/\sqrt{\alpha})}{\sqrt{\alpha}}]$$
(3.2)

This equation for the case $\sigma=0$ has been independently obtained by Jancovici (to be published, 12).

Figures 2 shows the mobile charge profile for various ratios of the background densities α_2/α_1 . The value 16 is scaled to represent the difference in density between a 1 molar electrolytic solution and a simple metal: If we take this number to be 64, then to go from three dimensions to two dimension, we simply take the $(2/3)$ power

which is 16. Clearly, most metals are far from being classical one component plasmas (Drude Theory), although semiclassical treatments are widely used to explain transport properties (14), which, as we know, are strongly dependent on correlations. Unfortunately the quantum mechanics of fully correlated systems is very difficult, more so in the vicinity of a surface. For this reason we believe that an exactly solvable model of a fully classical metal interface is useful because it illustrates the subtleties of the behaviour of correlations in charged systems. Figure 3 shows the total charge distribution near the interface as a function of the background ratio. Although it is clear that the penetration depth on the metal side is smaller for the 'metal' side (side 2), the charge density is also much larger. Figure 4 and 5 show the rather strong dependence of the charge profile on the surface density σ . In Figures 6 and 7 we have compared the charge profiles for different σ , but keeping σ constant. Again, we see the rather dramatic dependence of the charge profile on the surface charge. Figure 8 shows the dependence of the contact potential on the surface charge σ , compared to the ideal electrode case.

To investigate further this point consider Poisson's equation (15).

$$\nabla^2 \psi(x) = -2[\rho(x) - \alpha(x)]\pi - 2\sigma\delta(x) \quad (3.3)$$

where $\alpha(x) = \alpha_1$ for $x < 0$ and $\alpha(x) = \alpha_2$ for $x > 0$.

By elementary integration

$$\psi(x) = -2\sigma|x| + 2 \int_{-\infty}^x dx_1 (x-x_1) [\rho(x_1) - \alpha(x_1)] \frac{\pi}{4} \quad (3.4)$$

Using (3.1), we get after some straightforward calculation

$$\Delta\phi = \phi(\infty) - \phi(-\infty) = -(1/4) \ln(\alpha_2/\alpha_1) \quad (3.5)$$

This extremely simple formula for the contact potential of two metals (or, correspondingly, to the potential of zero charge for the case of the metal electrolyte interface), is not really unexpected when the charge $\sigma=0$: Actually this result was obtained on the basis of a

$$f_i = F_i / kT_N = -(1/4) (e_i^2 / kT) \ln \alpha_i + F_o(T) \quad (3.6)$$

where i is the index for the side of the interface ($i=1,2$).

The reversible work to transfer one mobile charge from side to the other is clearly

$$\Delta f = (-1/4) (e^2 / kT) \ln(\alpha_2/\alpha_1) \quad (3.7)$$

But the reversible work is only electrostatic, so that

$$\Delta f = (e^2 / kT) \Delta\phi = (-1/4) (e^2 / kT) \ln(\alpha_2/\alpha_1) \quad (3.8)$$

where we must remember that because of our special choice of units the electrostatic potential is measured in units of $2kT/e$

This result is contained in the work of Ballone, Senatore and Tosi (18) who showed for the neutral interface that the potential drop across the interface depends only on the difference of the chemical potential in the bulk phases.

The surprise is that the surface charge has no effect whatsoever on the potential drop across the surface. Clearly this must be due to the fact that because of the perfect screening theorems (16,19,20) the charge distribution has no multipole moment. Indeed the theorems have been proven for finite size disks (for R finite), and in that case $\langle x \rangle = 0$, which implies no potential drop due to charge σ . We conjecture that this relation remains valid in the limit of $R \rightarrow \infty$.

Just as an interesting aside, if we scale the relation (3.8) to three dimensions, then, since

$$\phi = n^{(2/3)} \quad (3.9)$$

$$\Delta\phi = (-1/6) \ln(n_2/n_1) + F(T) \quad (3.10)$$

where n_1, n_2 are the bulk electron densities of the metals, and $F(T)$ is

is a function of the temperature alone.

IV THE GAP MODEL

The equation (2.16) offers a rich variety of possibilities that we shall not explore at this time. We have plotted the charge density densities for two different cases in figures 9

and (10) (total charge density). More complicated profiles are obtained when the interface is charged and/or polarized. However, the more interesting quantity is the electrostatic potential, which is obtained by integrating Poisson's equation (3.3). The results for the charged gap where $\sigma_1 = \sigma_2 = \sigma$ and the background $\alpha_2 = 4$ are shown in Figures 11 and 12

again here we observe that the effect of the charges is screened out completely, and the potential drop is given by (3.5). We finally computed the case in which the interface had a permanent dipole by setting $\sigma_1 = -\sigma_2 = 2$. Figure 13 shows the two opposing orientations

of the dipole, just to show that they are also completely screened out. Figure 14 is the same calculation but with $\alpha_2 = 4$. Again here, the potential drop is given by (3.5). This is a consequence of the perfect screening theorems.

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FIGURE CAPTIONS

Figure 1 System geometry.

Figure 2 Mobile charge density profile $\frac{1}{2}\rho(x)$ Curve 1, $\alpha_2=1$; 2, $\alpha_2=2$; 3, $\alpha_2=4$

4, $\alpha_2=8$; 5, $\alpha_2=16$

Figure 3 Total charge profile $\frac{1}{2}\rho(x)-\alpha_2x$, same numeration of curves as Figure 2. No gap in this case; $\ell=0$

Figure 4 same as Figure 3 but with $\sigma=1$.

Figure 5 same as Figure 4 but with $\sigma=-1$.

Figure 6 Variation of total density profile with charge for $\alpha_2=2$: Curves 1, 2, 3, 4, 5 correspond to surface charge densities $\sigma=-4, -2, 0, 2, 4$

Figure 7 same as Figure 6 but with $\alpha_2=16$

Figure 8 Contact density as a function of the surface charge

Curve 2, $\alpha_2=4$; 3, $\alpha_2=16$

Figure 9 Total charge density profile for a gap model with $\ell=1$

Here both plates are equally charged Curve 1, $\sigma_1=\sigma_2=2$; Curve 2 no charge

Curve 3, $\sigma_2=-2$.

Figure 10 Same as figure 9, but with $\sigma_1=-\sigma_2=+2$, $\alpha_2=4$. Case with dipole

and background gap.

Figure 11 Potential profiles for the same cases as in Figure 9.

Figure 12 Same as figure 11, but with $\alpha_2=4$.

Figure 13 Interface with dipole, no background gap. Potential for same case as Figure 10 with $\alpha_2=1$.

Figure 14 Same as Figure 13, but with background gap $\alpha=4$. Corresponds
2
to the charge density of Figure 10.

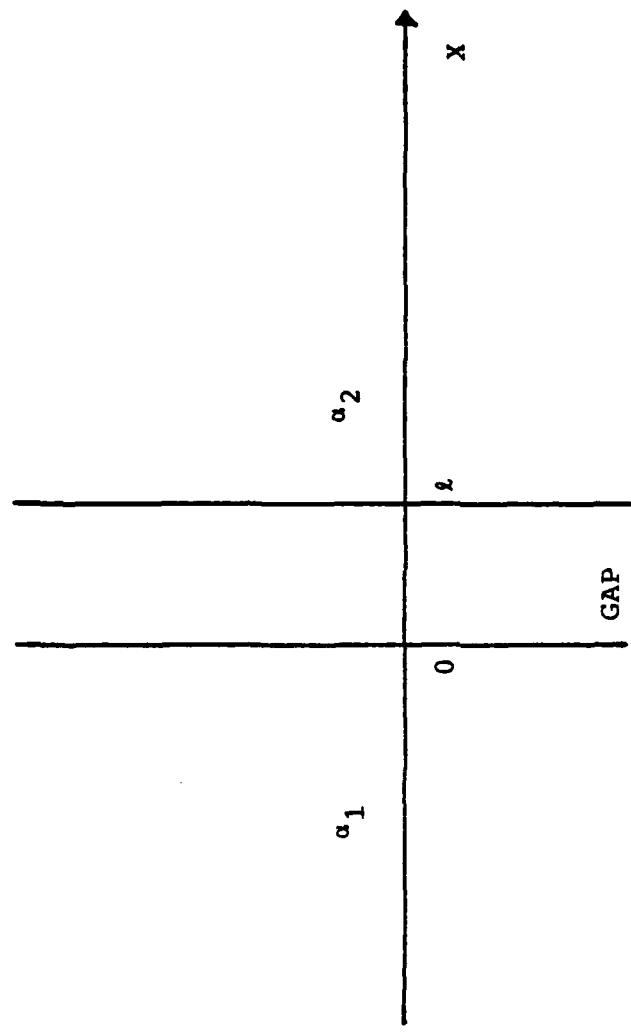


FIGURE 1

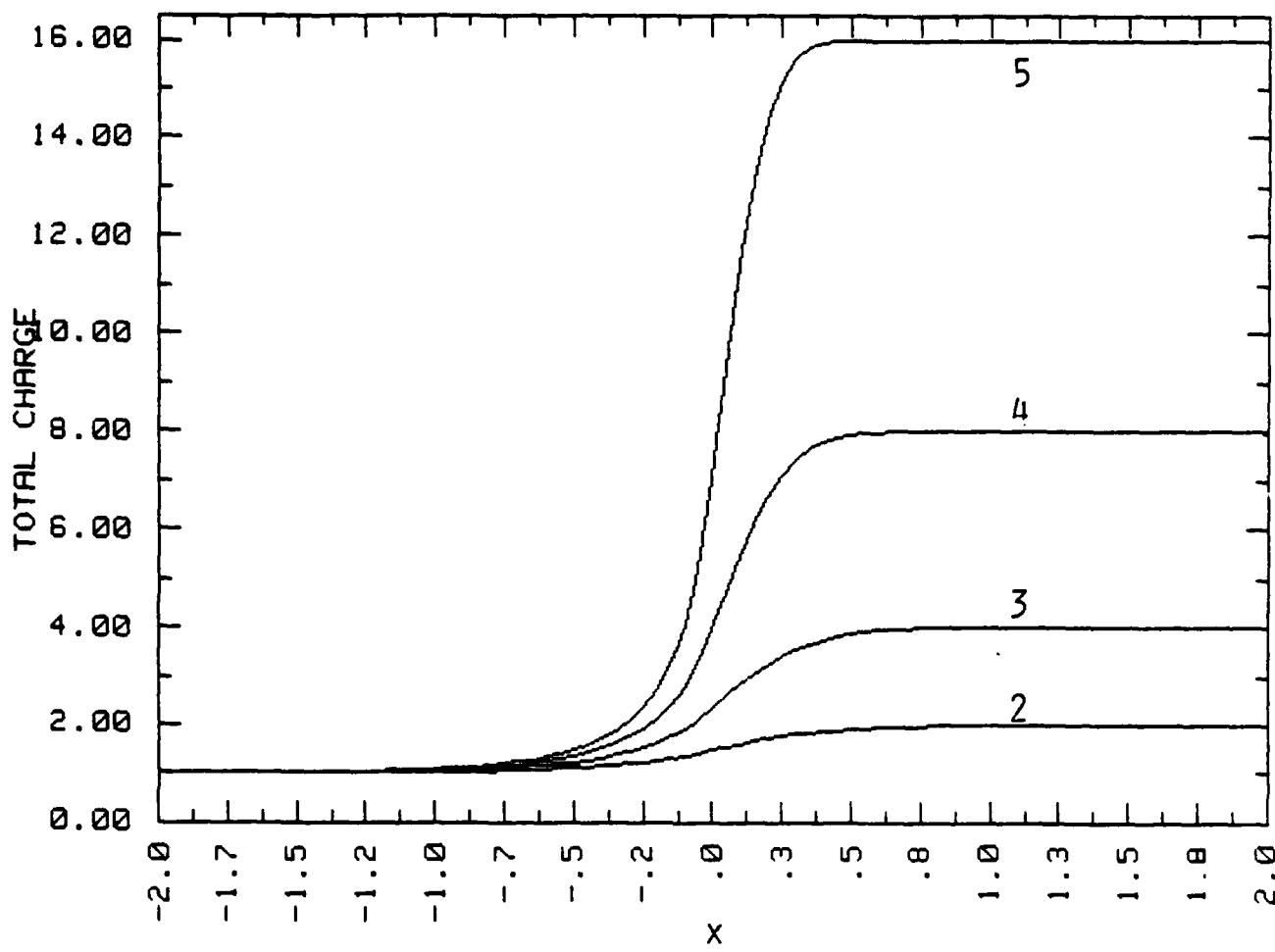


FIGURE 2

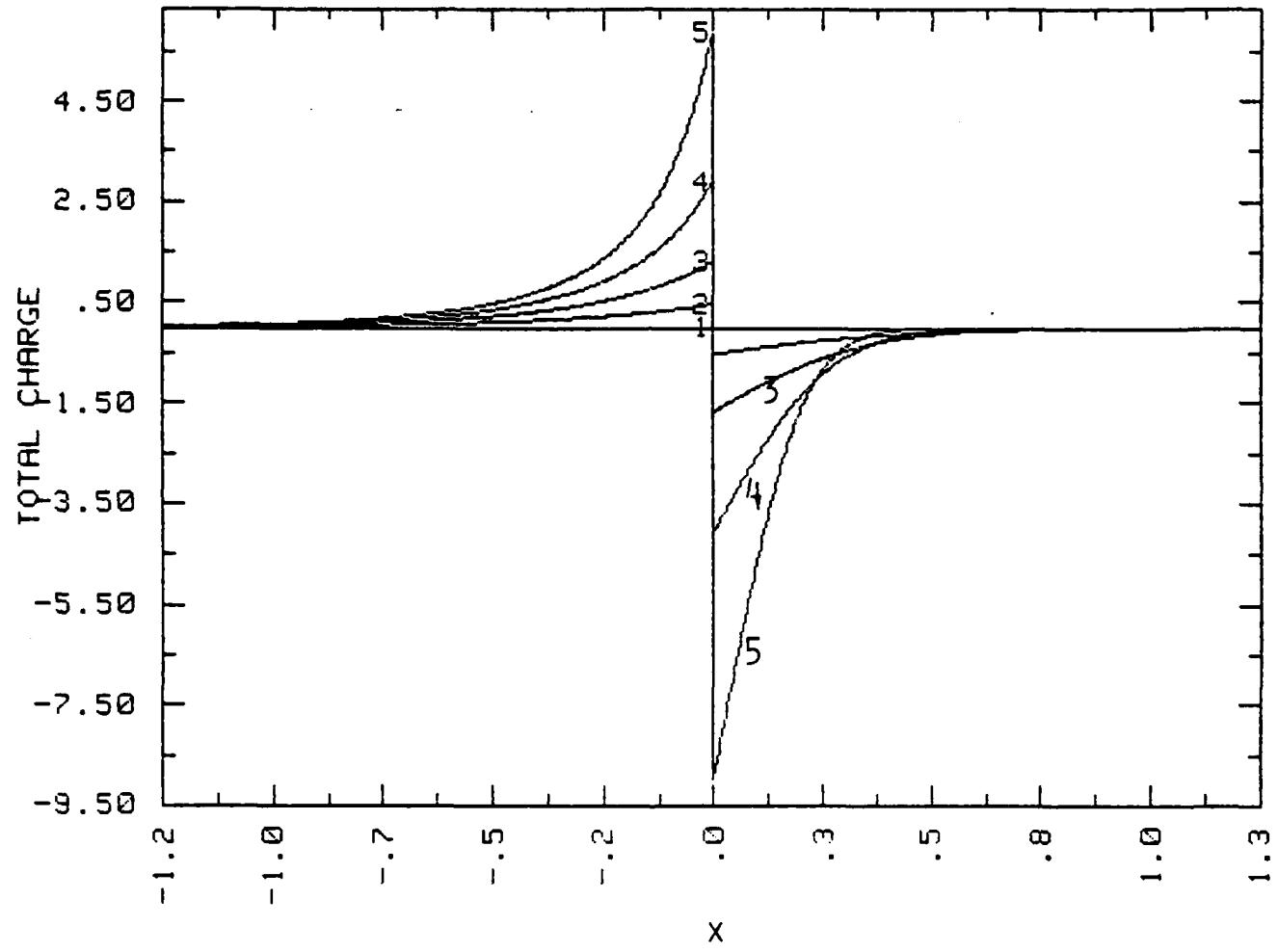


FIGURE 3

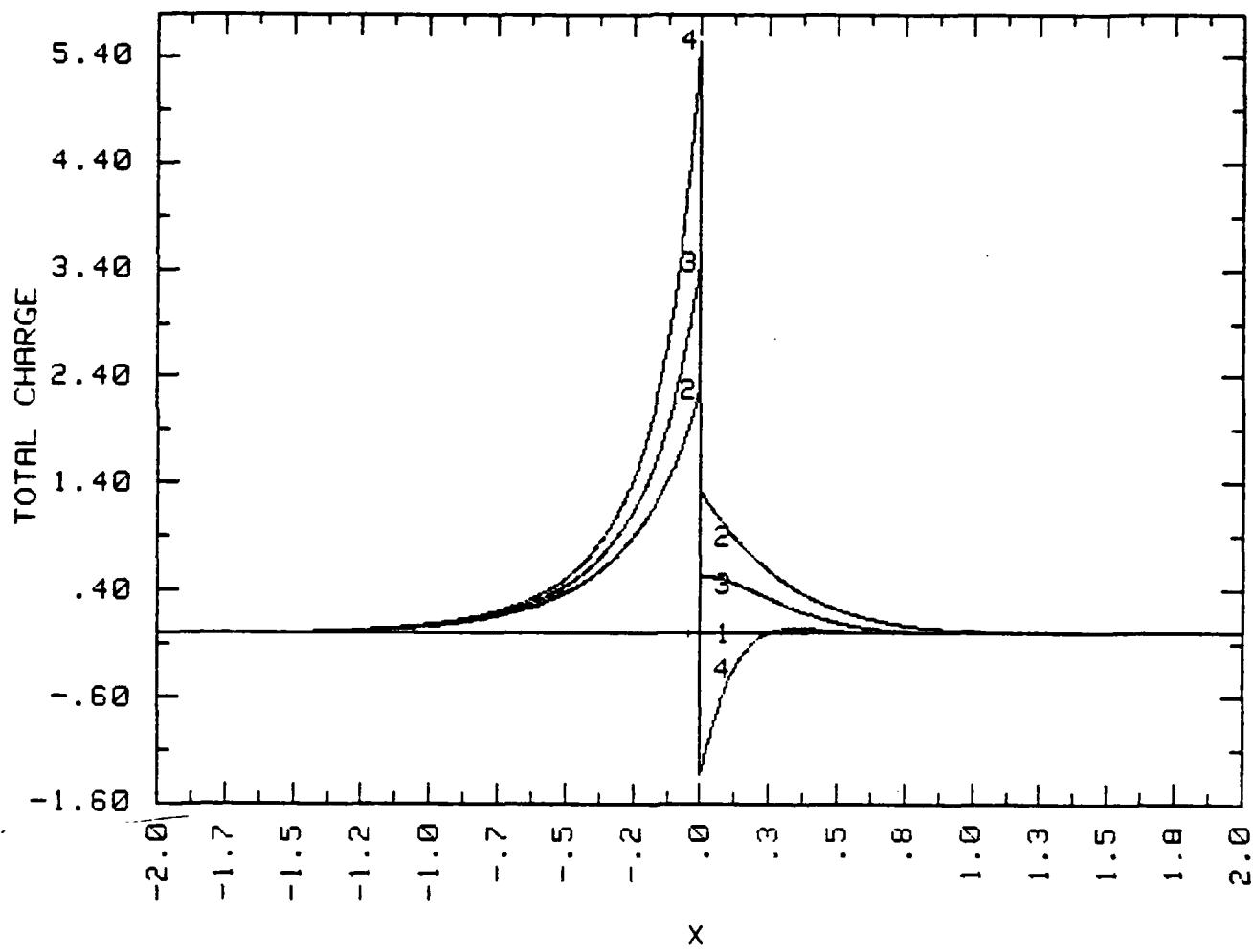


FIGURE 4

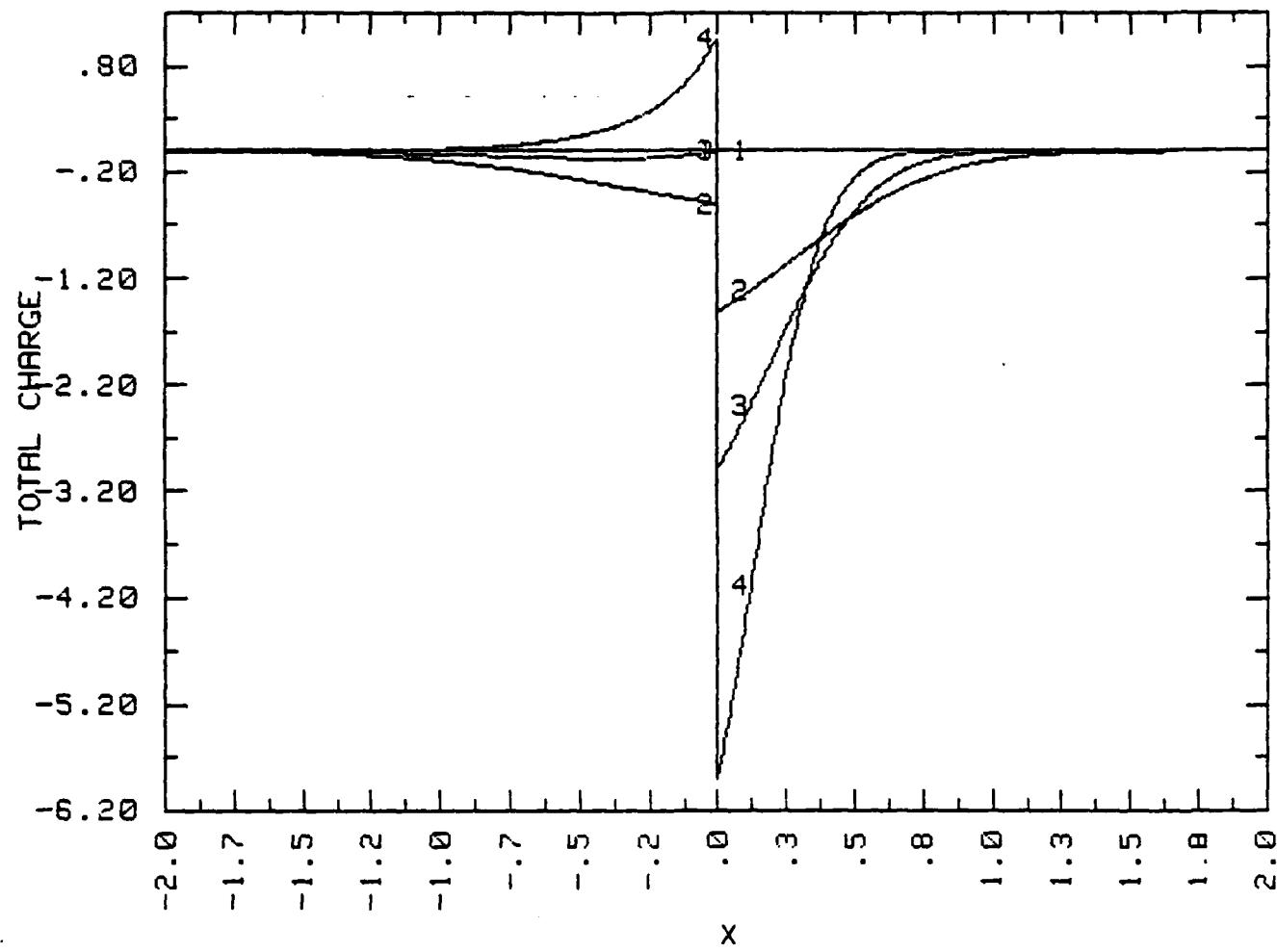


FIGURE 5

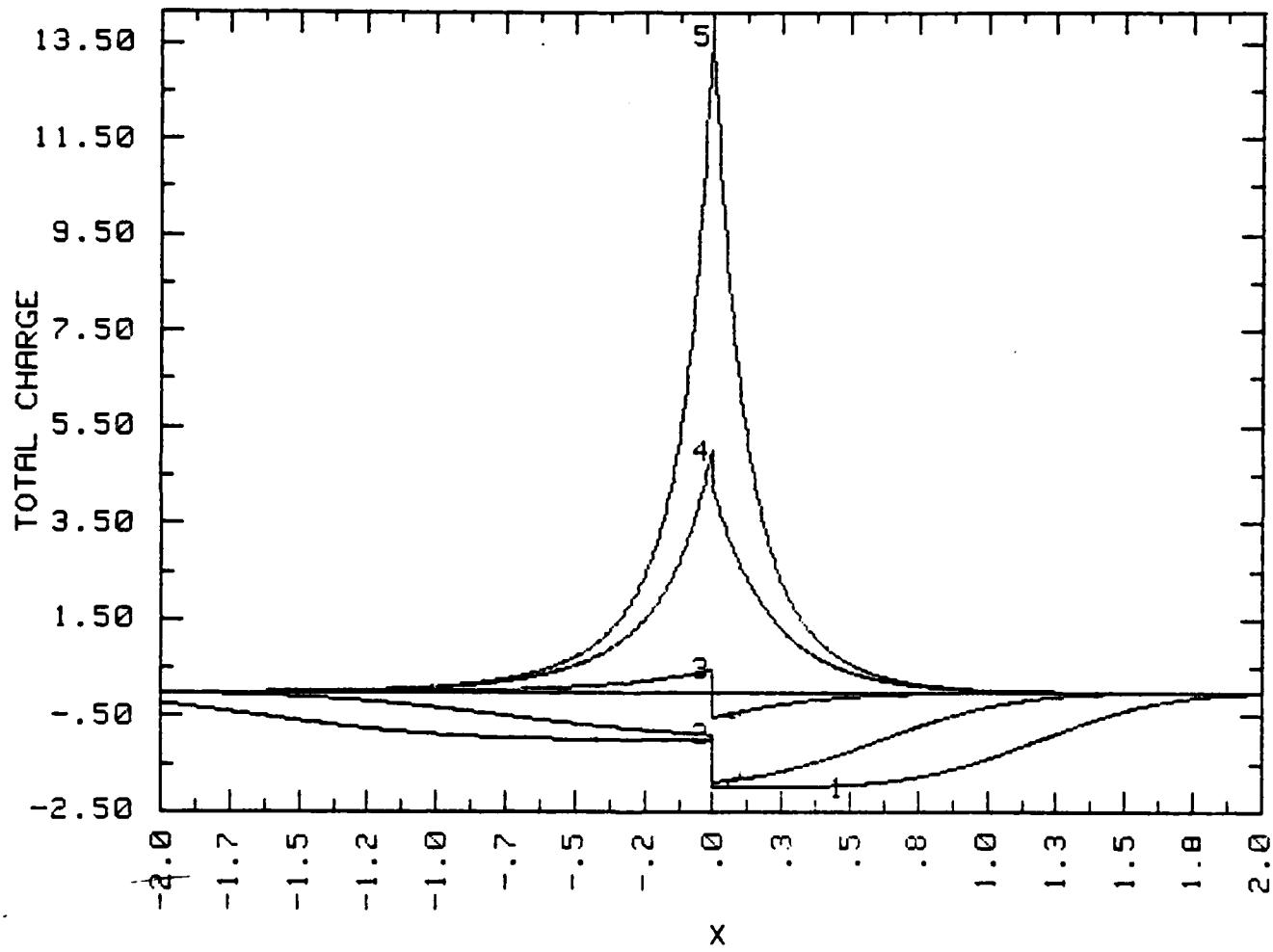


FIGURE 6

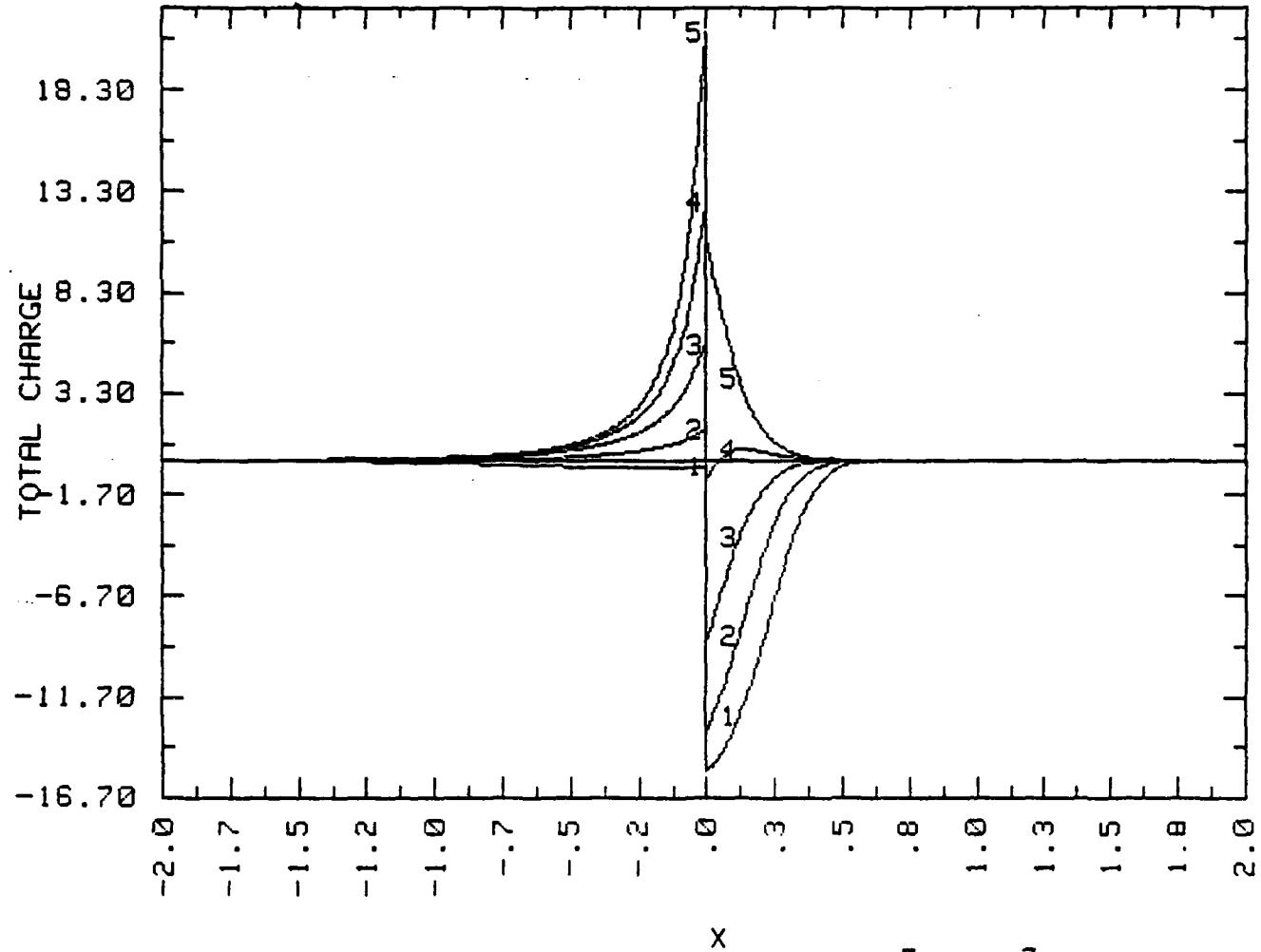


FIGURE 7

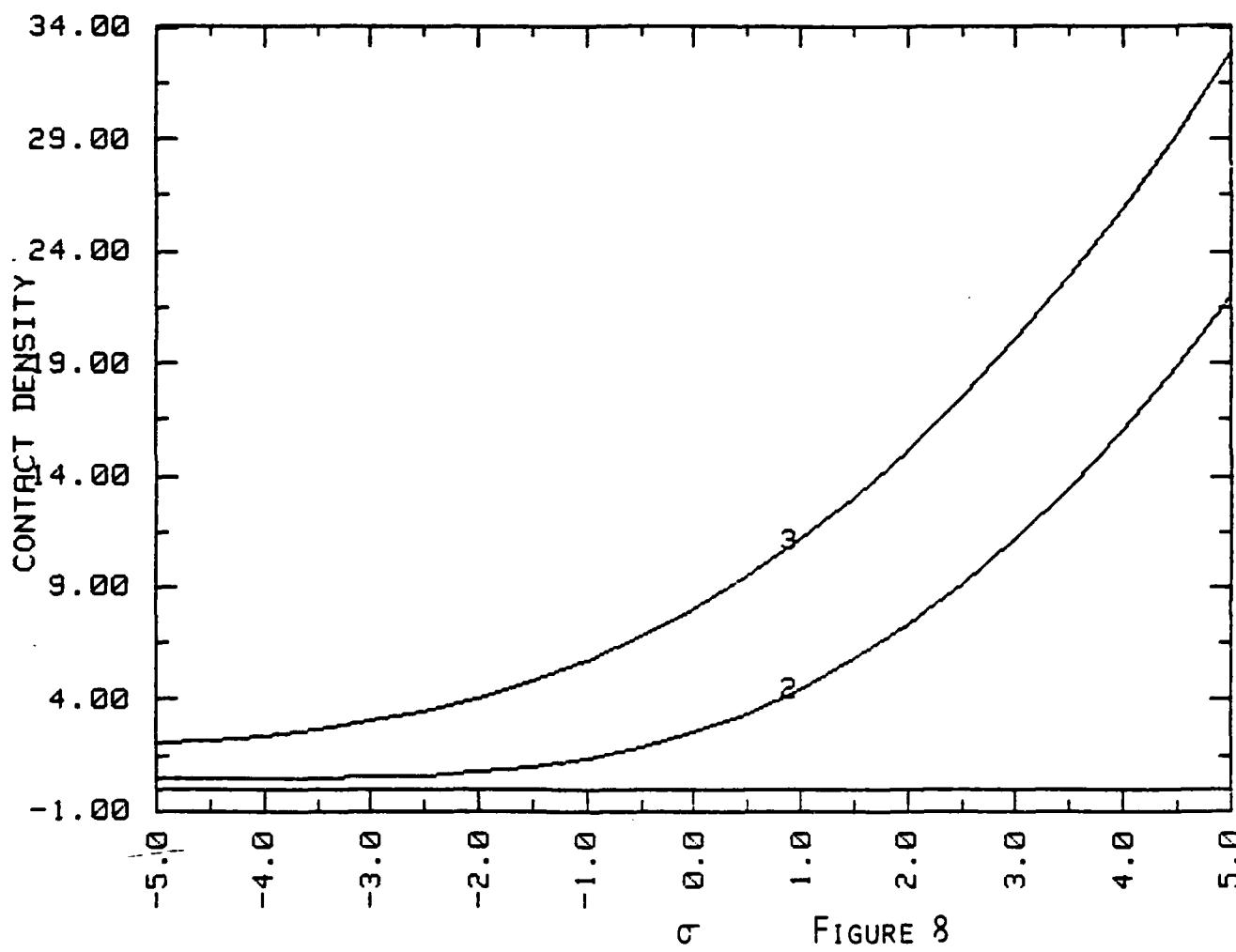


FIGURE 8

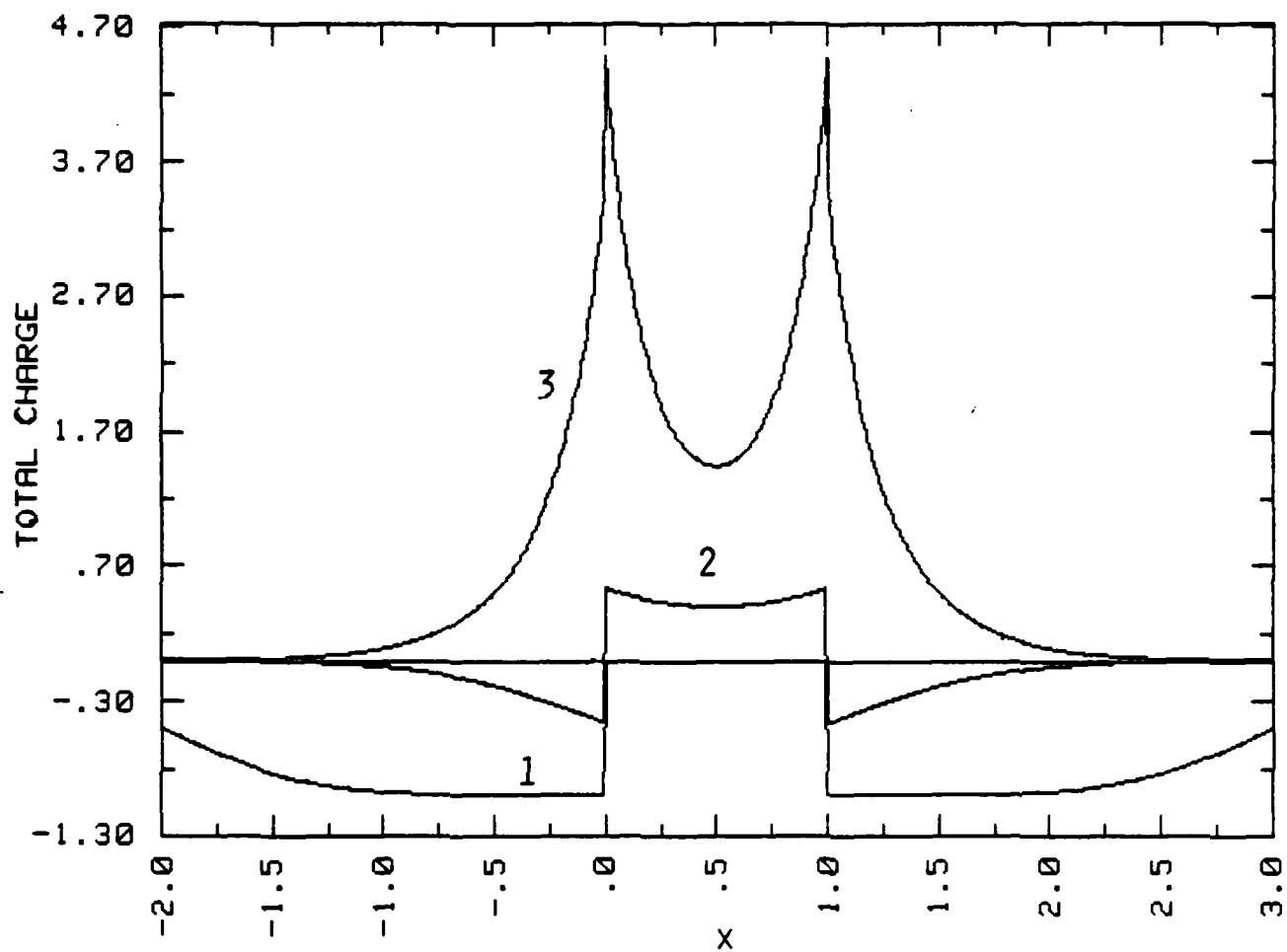


FIGURE 9

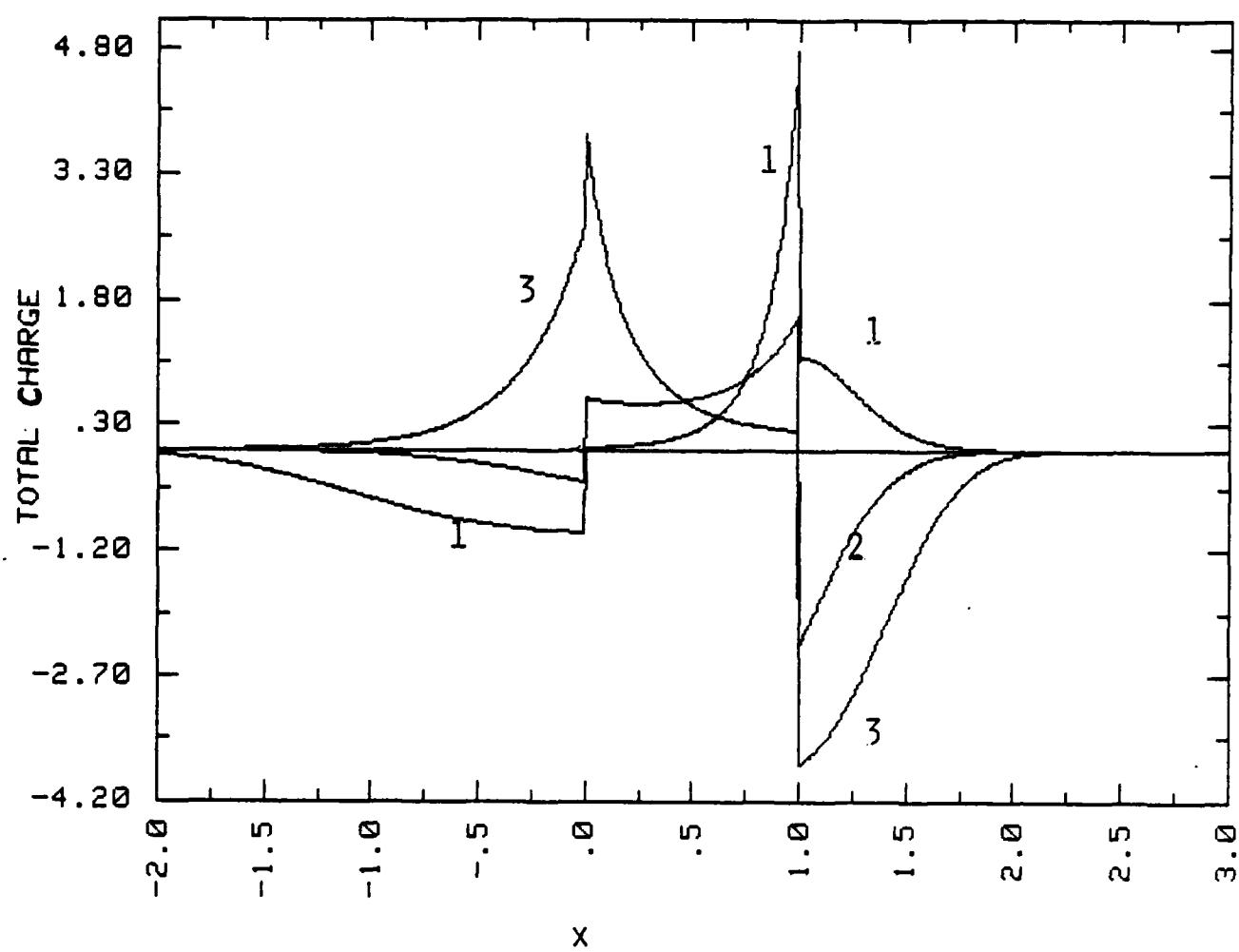


FIGURE 10

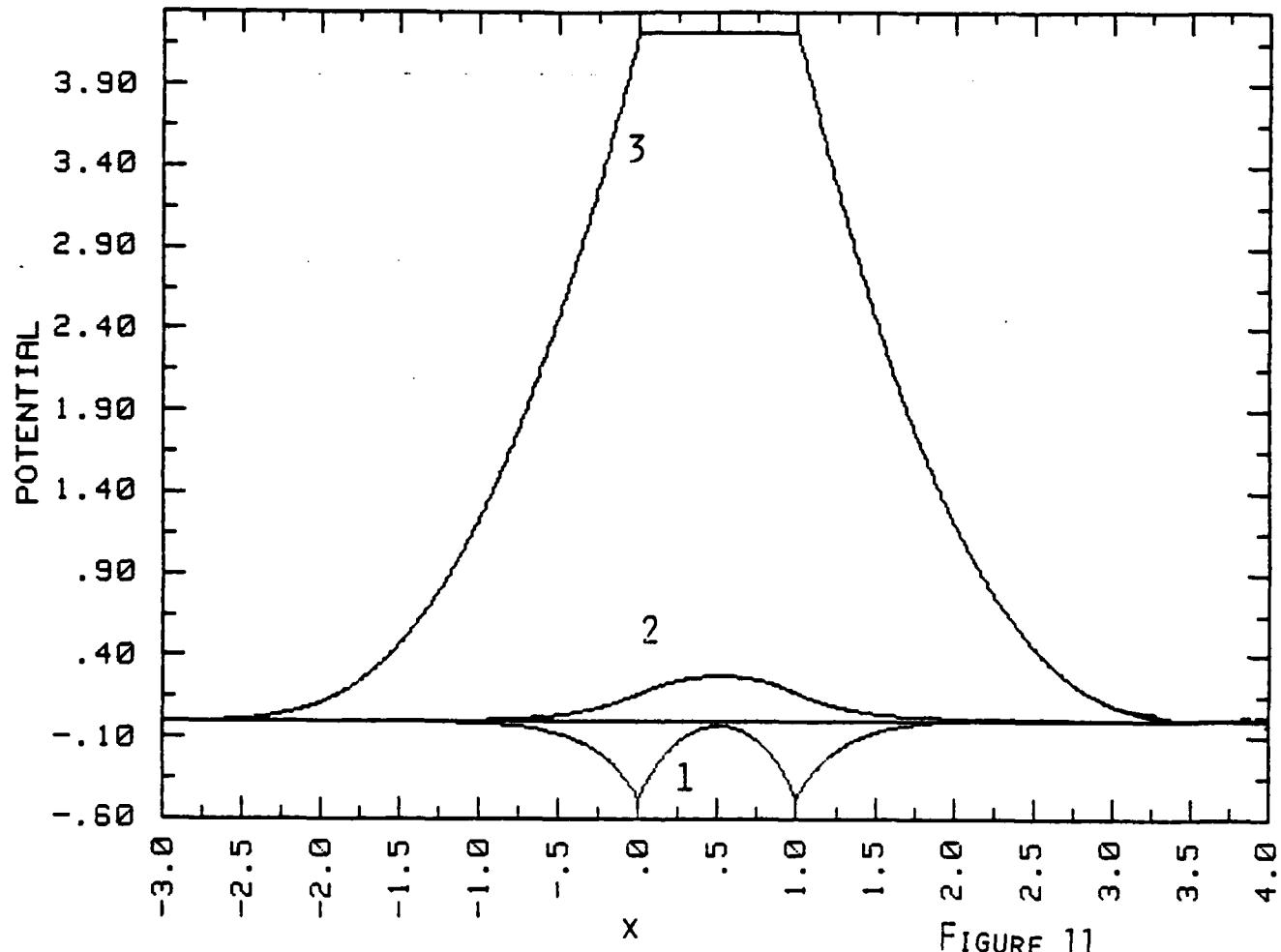


FIGURE 11

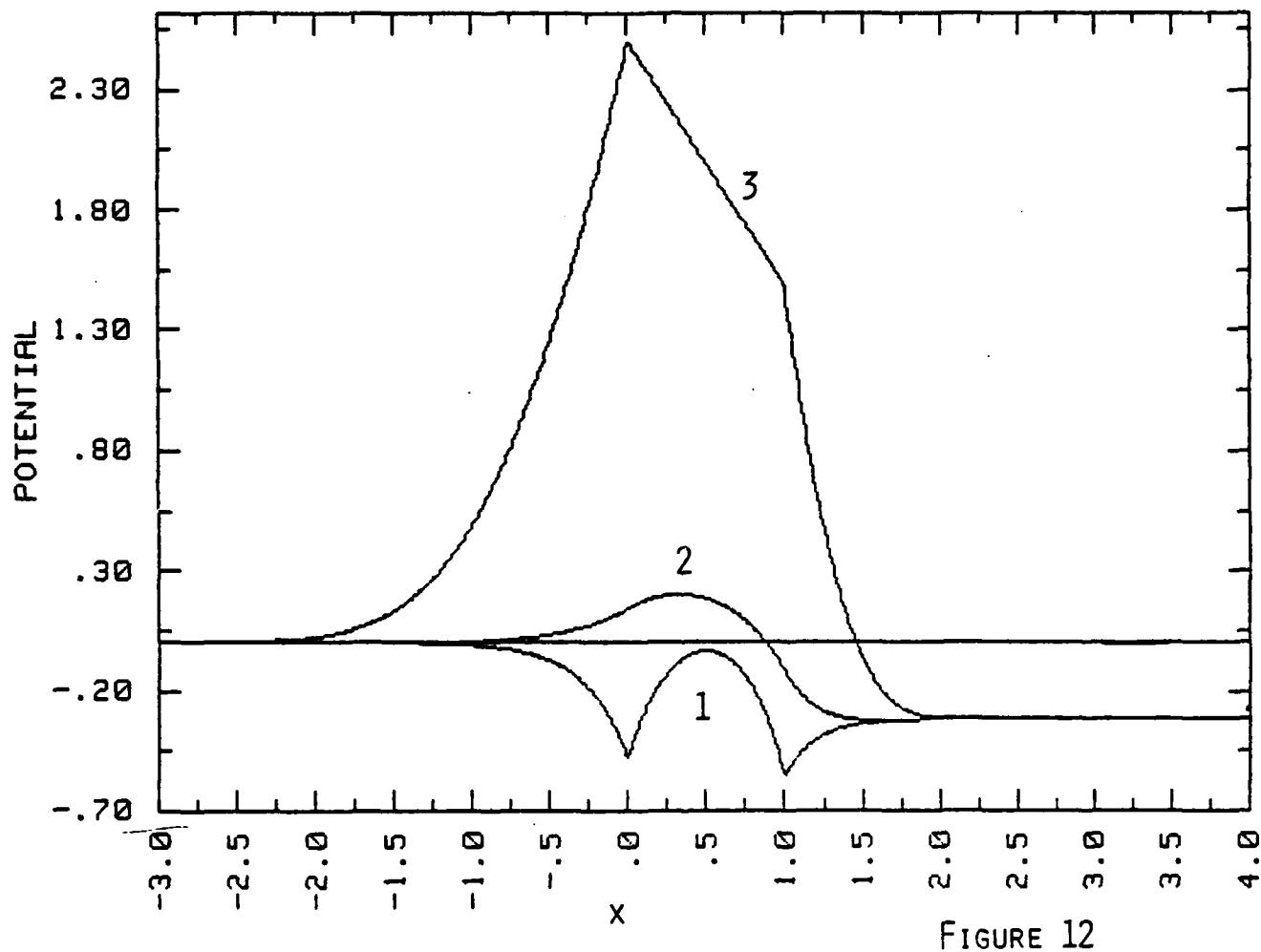


FIGURE 12

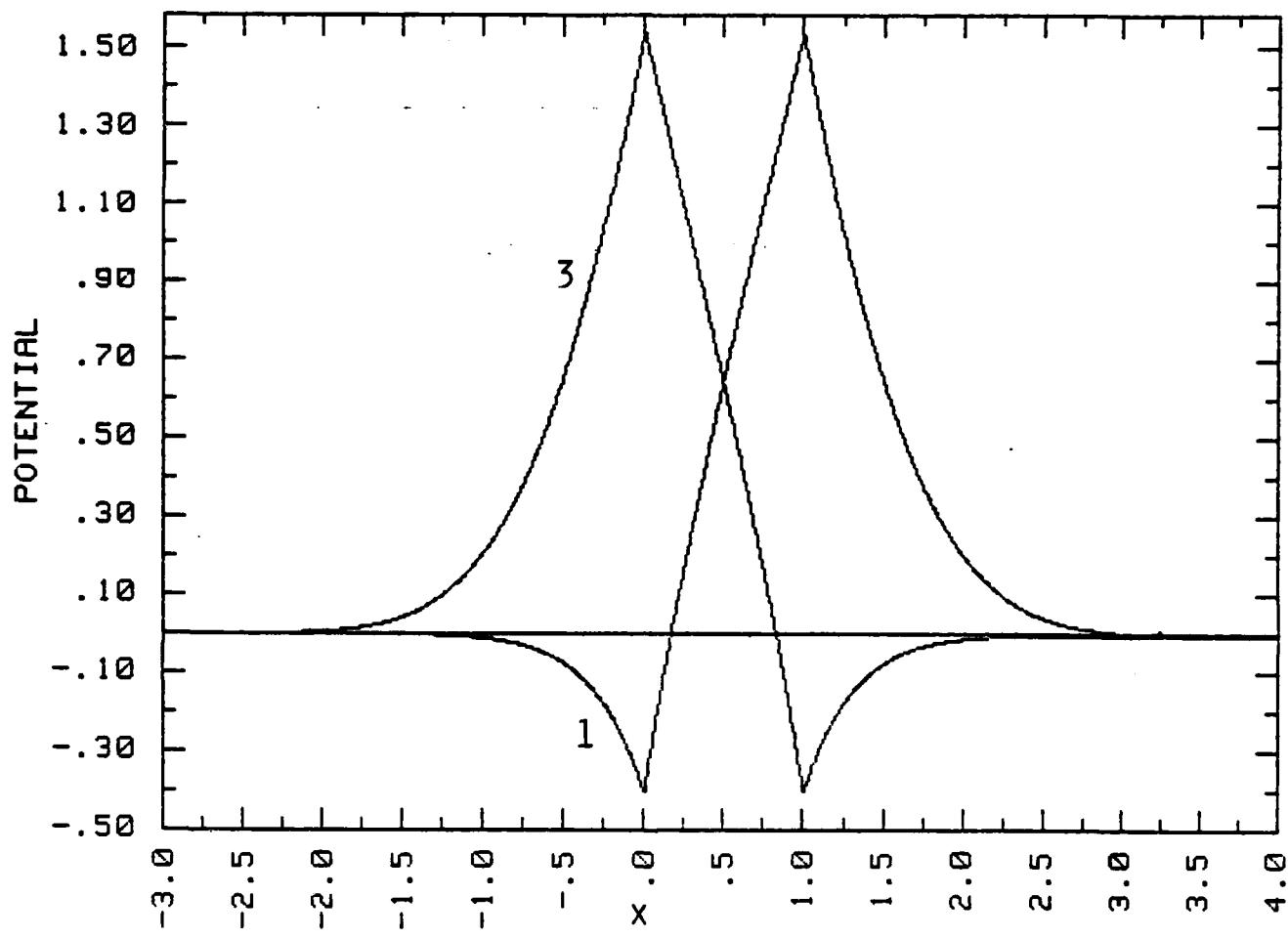


FIGURE 13

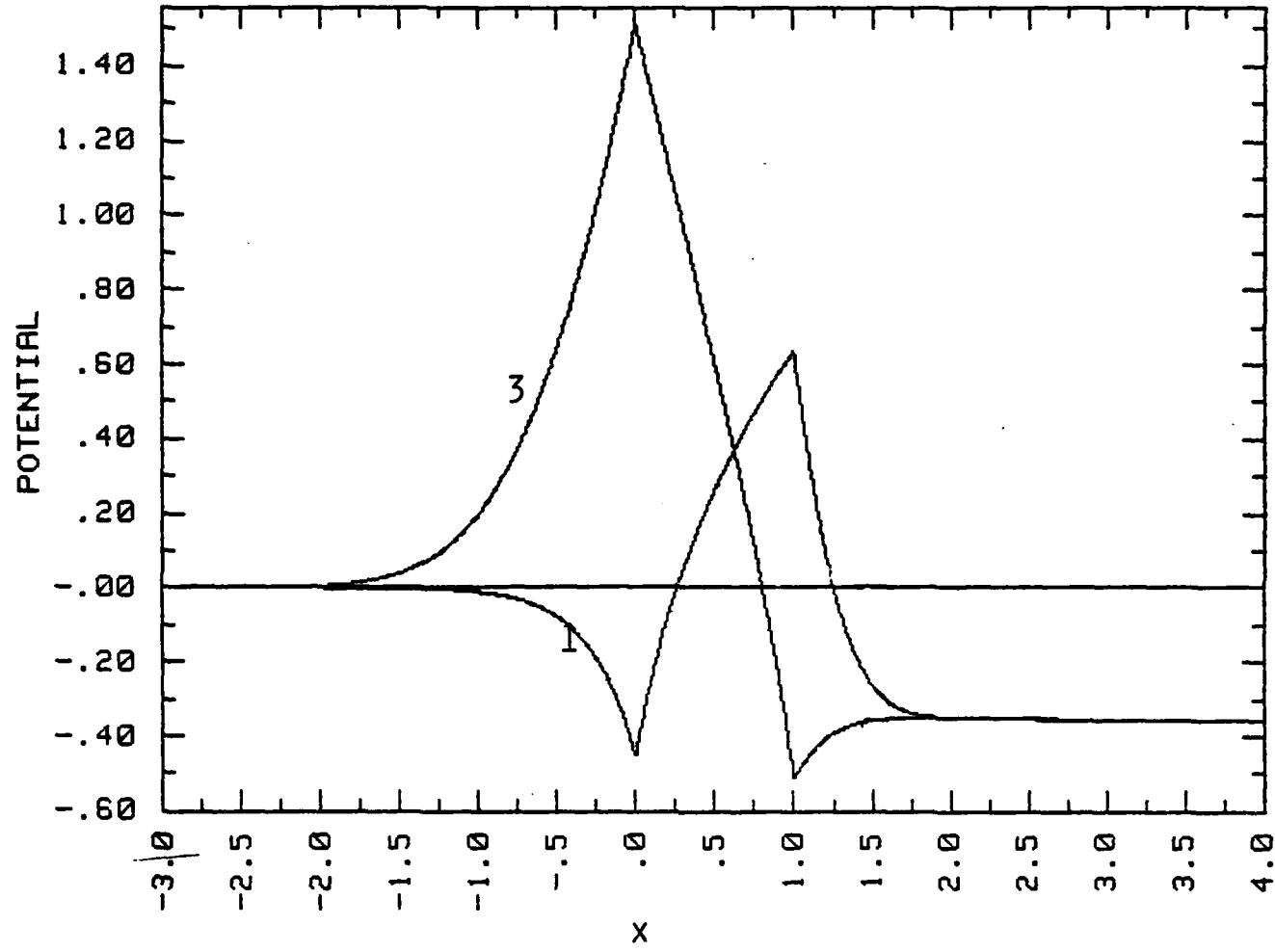


FIGURE 14